ARTIFICIAL GRAPHITE

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The sharp increase in graphite consumption by a number of industries requires the finding of new sources of graphite.

Development of new deposits takes too long (4-5 years) and requires considerable investment (at least 4 million rubles per 1000 tons of production). Besides, deposits of practical interest are unfavorably located at the periphery of the Union territory.

In the fall of 1935, the Institute of Mineral Raw Materials (IMS) attracted the attention of interested agencies to the necessity for detailed study of the problem of artificial graphite and in 1936 initiated research work on the subject.

Preliminary study of the problem revealed that production of artificial graphites may be organized considerably sooner, requiring an investment considerably smaller (1.5-2 million roubles per 1000 tons) than the development of new deposits of natural graphite; location of the plant for producing graphite depends only on the possibility of obtaining electric power.

Acheson Graphite

Works on Acheson graphite were initiated by the IMS in 1930, simultaneously by with a study of the method for thermal refining of natural graphites. At that time the subject of producing artificial graphite was absolutely obscure. There were no specialists on the subject in the USSR; there was no possibility of getting any adequate information from abroad. Data from world technical literature were also insufficient not only for the organization of production but even for judgment on the possibility of such production in the USSR.

Therefore, the investigators of that period had to find a complete solution of the following problems:

1. Development of the theory of coal graphitization, since data published on this subject are extremely contradictory and sometimes theoretically scholastic.

2. Study of graphitization procedure on an industrial scale: the process of graphitization, in spite of its wide discussion in scientific publications, is described superficially. This information may indicate only a possible trend of investigation.

3. Study of conditions for using artificial graphite: In this respect, there were only vague indications that artificial graphite is used in certain fields of industry.

To solve the first problem, it was necessary to start from the universally adopted chemical theory, which maintained that transformation of amorphous carbon to graphitic carbon occurs in the graphitization process under the effect of heat. Essential role in this theory was played by the concept of catalytic action of ash admixtures.

After two years of investigations, the IMS positively established that dispersed structure of raw material is of major significance for graphitization and the properties of its products. Mineral admixtures play a quite secondary role, and the graphitization process itself has to be considered as thermal recrystallization of colloidally-dispersed graphite already present in coal.

Thus, the physicochemical theory of recrystallization was accepted instead of the completely rejected chemical theory.

To study the technique of graphitization, experiments were arranged at the Tsaritsyn experimental plant of the Institute and later at the "Elektrougli" plant.

1. To investigate the general economies of the problem, compiling a balance of graphite production and consumption in the Union; particular attention had to be paid to fine-crystalline graphite, scarcity of which is of a most threatening character. To establish possibilities for eliminating the deficiency, and the role of Acheson traphite in this respect.

2. To improve the technology of graphitization under industrial conditions, using the experience of the "Elektrongli" and Moscow Electrode plants.

3. To prepare large samples of artificial graphite under entirely definite conditions and to lest systematically their applicability in various branches of industry, chiefly in the cell, pencil and lubrication productions.

4. To test the samples of artificial graphite in various branches of industry and to establish conditions for its use without confining to already existing methods, developed for natural graphites.

For fulfilment of these tasks, the following projects were carried out:

1. Experiments for graphitization at the Moscow Electrode Plant - 2 campaigns.

2. Experiments for graphitization at the "Elektrongli" Plant - 6 campaigns.

3. Laboratory investigation of machining artificial graphite.

4. Preparation of samples for testing in cell and pencil productions.

5. Testing various grades of artificial graphite for manufacturing lubricants and graphite-colloidal compounds.

6. Further development of the theory of the graphitization process under industrial conditions.

7. Compilation of requirements for designing a plant for artificial graphite.

8. Initiation of planning a plant at Shakhty (Donbas).

Experiments at the Moscow Electrode Plant

These experiments were conducted in the furnace designated for graphitization of electrodes: distance between furnace electrodes 480 cm, cross-section of electrode packs 163 x 40 cm. The equipment happened to be in unsatisfactory condition, which had a negative effect on results as will be shown further.

Power of reducing substation amounted to 1500 kva, however the furnace was receiving an average 900 kw due to very low cos φ , about 0.6.

First campaign was conducted with graphite from Zaval'ye deposit; 5618 kg of 100 mesh graphite with 12% of ash content comprised the furnace charge. This material had very small loose weight, 0.62, and high electric conductivity, both factors being unfavorable for the process.

The furnace with 188 sq dm cross-section of working space was fed with current for 21 hours 10 minutes. Power distribution in installation was as follows:

	Kwh	%
Losses in potential regulator	213	1.15
Losses in transformer	677	3.64
Losses in bus-bars	360	1.93
Losses in electrodes	4,180	22.50
Total losses	5,430	29.22
Useful power	13,170	70.80
	18,600	100.02

The furnace yield amounted to 1933 kg of grade I product and 2975 kg of grade III.

During furnace unloading, samples of product were taken from the points located by a grate plotted on the cross-section of working space. Results of determination of the ash content in these samples are presented on the topogram in Figure 1, which shows that the very upper 25-cm-thick layer of graphite was not heated extremely high and remained practically unchanged. Under this layer there is a crust of grade III product about 25 cm thick. The ash content in the crust somewhat increased due to precipitation of vapors from the underlying grade I.. Formation of crust facilitated removal of grade I product from the furnace; cross-section of volume occupied by grade I was 60 x 103 cm.

Power consumption: 2) 3.31 kwh, i.e. approximately 10% higher than normal, per kg of charged graphite for entire installation and 2.34 kwh/kg fed into the furnace; b) 9.6 and 6.8 kwh/kg respectively for grade I product.

Increased specific power consumption may be explained by the impossibility of heating the furnace to a sufficiently high temperature due to abnormally high losses of power.

Analysis of the results of the campaign revealed that, in addition to losses in the circuit, amounting to 29.2%, considerable losses were sustained due to electric conductivity of the fused bottom of the furnace. These losses can not be precisely determined, but an idea of their extent may be obtained from the following considerations: according to a formula given below, the amount of power, required for heating the furnace, must be equal to 13,400 kwh; despite the fact that the furnace received this amount of power, it was not heated sufficiently high and yielded a decreased quantity of grade I product, 1933 kg instead of the expected 3370 kg; hence, power losses in the furnace hearth may be estimated at 30%.

Taking into consideration defects of equipment, the power supply for the next furnace campaign was $1^1/2$ times higher.

The second campaign was conducted with Donets anthracite roasted at 1300°C. Its main mass passed through 15-mesh sieve and the amount of fine fractions was very low; ash content was 3.5% and loose weight - 1.06.

The peculiarity of the material for processing in the given case lay in its coarseness in the absence of fine fractions, which factor always results in increased consumption of power.

Charge of anthracite was 5400 kg. The working space of the furnace had an almost cylindrical shape with average radius of 59 cm and cross-section of 109.5 sq dm. Current was supplied for 31 hours 40 minutes. Average cos φ = 0.59 on high side; average power on low side was 875 kw

Power distribution was as follows:

	Kwh	%	%
Losses in potential regulator	159	0.59	
Losses in transformer	954	3.45	
Losses in bus-bars	377	1.36	
Losses in electrodes	6,400	23.10	
Total losses	7,890	28.5	
Useful power	19,810	71.5	
	27,700	100.00	

Obtained 1,400 kg of grade I product and 3,000 kg of grade II.

Samples were taken during unloading; their ash contents are given on the topogram of Figure 2, which shows that the working space attained the required temperature only at its lowest part, about 40 cm from the bottom. The volume of Grade I

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Samples were taken during unloading; their ash contents are given on the topogram of Figure 2, which shows that the working space attained the required temperature only at its lowest part, about 40 cm from the bottom. The volume of Grade I this time is still smaller than in the previous campaign, despite a 1.5 - times greater consumption of power. This is mainly caused by excessive grain coarseness of the anthracite and high losses of power. More detailed analysis of the results of this campaign is of no interest since it deviates too far from normal results.

Conditions of experiments are summarized as follows:

1. Very high losses of electric power in installation, up to 39% in the end of heating and about 29% average for the entire campaign. Particularly high are the losses in furnace electrodes. These losses are mainly caused by too small a cross-section of the electrode pack and unsatisfactory design of electrode holders; the latter causes nonuniform distribution of current along the electrode cross-section.

2. High and undeterminate losses due to electric conductivity of furnace bottom.

3. High reactive resistances of installation resulted in low cos . This is mainly conditioned by design of the furnaces, which form a large loop with the bus-bars. Substantial significance is also attached to the high reactance of the transformer, equal to 17% at the lower stage.

Due to the defects listed above, further experiments had no sense.

Experiments at the "Elektrougli" Plant

Conditions for these experiments were incomparably more facorable than those at the Moscow Electrode Plant. Experiments were carried out in 250 kva furnaces with reactance about 0.0028 Ω and high cos φ , about 0.87. Six campaigns with various materials were conducted. Results are given in Table 1.

The initial resistance of the furnace is very high in the case of graphitizing finely divided anthracites, resulting in inefficient prolongation of the campaign. However this inconvenience may be eliminated easily by introducing a core of already roasted grade III anthracite, as was done in campaigns No 2 and No 6.

Donets anthracites from the Vlasov and Voykov deposits are very calm during heating. Heating conditions improve noticeably with a decrease in coarseness.

Ural anthracite, due to its high ash content and large amount of volatiles, shows considerable peculiarities in its behavior during graphitization. These peculiarities, requiring special study, are: 1) intensive and violent gas evolution in the beginning of the process, accompanied by explosions of oxyhydrogen gas and gushing; 2) tendency of anthracite from Brady deposit to cementation during graphitization into a solid sandy mass. Both peculiarities are accompanied by reduction of furnace resistance and by a change in thermal and electric conditions of the process for the worse; and they also considerably complicate unloading.

Table 1

Conditions of Experiments

	Campaign			
	1 - 2	3	4	5 - 6
Materials	Anthracite from Vlasov Deposit	Anthracite from Voykov Deposit	Graphite from Zaval'ye Deposit	Anthracite from Bredy Deposit
Loose Weight	0.71	0.9	0.65	0.63
Ash Content, in %	7.60	4.2	12	9.7
Volatiles	5.1	3.63	-	6.4
Screen Analysis (%):				
+ 50 mesh	-	0.18	-	-
+ 100 mesh	8.95	4.14	-	-

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Materials	Anthracite from Vlasov Deposit	Anthracite from Voykov Deposit	Graphite from Zaval'ye Deposit	Anthracite from Bredy Deposit
+ 150 mesh	0.42	25.6	-	5.7
+ 200 mesh	25.25	56.5	-	17.7
- 200 mesh	58.70	12.43	-	76.6
Dust	6.70	1.33	-	-
Working Space:				
length, in dm	21.5	21.5	27	21.5
cross-section, in dm	7.5 x 6.3	7.5 x 5.6	34.4	7.5 x 5.5
weight of material, in kg	700	650	600	470
weight of fill, in kg	2,500	2,500	-	2,500
Resistance, initial	-	2,280	75	600
Resistance, terminal	3.7	4.4	4.0	-
Voltage of Generator, in V:				
initial	170	187	135	175
terminal	32.2	35	34.5	33.5
Power, average, in KW	243	136	219	150
Furnace Voltage, in v:				
initial	154	184	118	172
terminal	16.5	19.5	14	19.5
Power Consumption:				
Total, in kwh	4,940	3,240	5,880	3,710
Distribution, in % :				
motor and generator	17.8	18.0	19.7	18.2

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Conditions of Experiments	Campaign			
	1 - 2	3	4	5 - 6
Materials	Anthracite from Zlasov Deposit	Anthracite from Voykov Deposit	Graphite from Zaval'ye Deposit	Anthracite from Bredy Deposit
bus-bars	25.3	2.3	36.3	23.8
electrodes		20.8		
furnace	56.9	58.9	44.0	58.0
Unloaded from Furnace, in kg:				
grade I	420	285	240	280
grade III	163	120	240	94
Losses, in kg	127	245	120	96
Specific Consumption of Power, in Kwhr/kg:				
Per kg of Charge:				
total	6.95	5.0	9.8	7.9
without losses	3.95	2.94	4.3	4.58
Per kg of Grade I:				
total	11.8	11.4	24.5	13.25
without losses	6.7	6.7	10.8	7.65

Therefore, special investigations are necessary for establishing the graphitization procedure for Bredy anthracite.

Campaign No 4 was conducted with 100-mesh Zaval'ye graphite. Initial resistance of this graphite is considerably lower than that of anthracites, but terminal resistance is just a little lower; this causes unfavorable distribution of current and temperature in the working space, resulting in increased power consumption.

In conclusion, it should be noted that the graphitization installation of the "Elektrougli" plant has excessively high electric losses, 45 - 44%, out of which 18 - 20% relate to the motor-generator and 20- 35% are attributed to furnace electrodes.

Curious conclusions are obtained from a comparison of the specific power used by a furnace upon charging it with various materials.

For materials with high initial electric resistance, such as Zaval'ye graphite and coarse roasted anthracite, the specific power consumption is sharply increased.

In the case of using materials close to each other in their properties, power consumption per kg of grade I product does not depend on the total power received by the furnace. Certainly, this relates only to the same furnace and fine powders. In spite of some failures, the experiments fulfilled the given requirements: all necessary design data were obtained and artifical graphite was produced in an amount sufficient for an investigation of its applicability.

Laboratory experiments. It was known that the quality of graphites obtained from different anthracites is not the same, that certain anthracites give artificial graphite particularly soft and fat to the touch. But the cause for this phenomenon remained unclarified and, therefore, proper selection of an optimum raw material for graphitization was impossible.

To solve this problem, several samples of anthracites, as far as possible different and typical in their character, were studied comparatively under conditions of graphitization. Results obtained are given in Table 2.

Table 2

Anthracite Brand	Ash Content, %	Volatiles, %	Anthracite Brand	Ash Content, %	Volatiles, %
Steklyannyy	1.7	-	Voykovskiy	4.2	3.9
Mochalinskiy	3.5	-	Bredinskiy	9.7	6.4
Grushevskiy	9.8	-	Poltavskiy	10.71	-
Vlasovskiy	7.6	5.1	Mikhaylovo-Leont'yevskiy	4.82	-

Steklyannyy, Mikhaylovo-Leont'yevskiy, Mochalinskiy and Grushevsk anthracites had close texture with a glassy shell-like fracture and gave sturdy sand-like graphite. Vlasovskiy anthracite produced soft and fatty graphite, while Voykovskiy anthracite gave graphite with intermediate properties. All these Donets anthracites in general give similar graphites differing from each other only by the thickness of plates in swelling (in graphitization of large lumps) and, consequently, by fatness and softness.

Graphite out of South-Ural anthracites, such as Bredinskiy and Poltavskiy, considerably differ from graphite made of Donets anthracites. It is less silvery and considerably softer, being easily pulverized.

Investigation of the structure of anthracites permits establishing some rules for proper selection of anthracite. Fatty graphite is obtainable from anthracites with fine-cleavage structure. Compact anthracites with glassy shell-like fracture give sturdy sand-like graphite.

It was noted earlier that more ashy anthracites generally give softer graphite, and attempts were made to explain this by the catalytic action of ashy admixtures. The incorrectness of this assumption was established by the author as long ago as 1933. Experiments permitted explaining the significance of ash content by the fact that finely-laminated anthracites, due to conditions of their genesis, generally have a higher ash content. Thus; the increased ash content may serve as a feature favorable for graphitization only because it is connected with a finely-laminated structure.

Anthracites from the South Ural belong to the finely-laminated type. In addition, X-ray examination shows that lameblar crystals of graphite in these anthracites are oriented parallel to the plane of cleavage; this also favors graphitization, and therefore these anthracites yield especially soft graphite. Their substantial deficiency from the technological viewpoint is a high ash content and large amount of volatiles. This factor not only decreases the yield of graphite, but also hampers heating of the furnace due to intensive gassing.

Further it was found that the properties of graphite depend, to a great extent, on the grain size of the initial anthracite powder: the coarser the anthracite powder subjected to graphitization, the fatter and softer is the graphite, other conditions being similar. The explanation is that large lumps offer more favorable conditions for distortion of the material and its splitting into thin laminae.

The mechanism of anthracite swelling during graphitization is still not clear. The cause for swelling appears to be a distortion due to heterogeneity of the material which forms the snthracite, whereupon a lump splits into more or less thin laminae and swells like a puff pasty. Stresses required for swelling and distortion are more easily induced in large lumps, while swelling may not occur at all in small parrticles, since an entire granule may be distorted as a whole without lamination.

Finally, the significance of mechanical treatment of graphitized anthracites was clarified. Acheson graphite, immediately after graphitization, possesses a certain rigidity and, compared with natural phanero-crystalline graphite, low slipperiness and fatness. Therefore, it is not plastic, can not be shaped by pressing and does not improve the forming capacity of the material to which it is being added. This factor prevented the penetration of artificial graphite into various branches of industry.

Experiments for grinding artificial graphites of varied origin in various types of mills established that all graphites become softer and fatter if the particles are crushed in the grinding process. Steel-ball mills are most effective in this respect. When they are used, differences in the properties of graphites from different initial materials are noticeably obliterated. However, other conditions being similar, a coarser initial material gives a fatter product. Simultaneously, the pulverizing effectivness of a mill decreases.

Thus, in the mechanical treatment of graphite it is necessary to vary its grinding and "polishing". The phenomenon of polishing is conditioned by crushing graphite grains, whereupon lamellar crystals in grains obtain parallel orientation and the grains themselves split into thin laminae. Such a deep modification in the dispersed structure of the material occurs sometimes with unusual facility. For example, Zaval'ye graphite, having comparatively thick crystals, after only slight crushing, acquires a fine-lamellar structure and becomes extremely fat. It should be noted that screen analysis almost does not show any pulverization in this case.

Fine powders of artificial graphites are much more difficult in polishing. But evidently, sufficiently intensive polishing may attain the necessary fatness in this case also. On the other hand, grinding of the initial anthracite (and, in addition, graphitization in furnaces) is incomparably easier than the grinding of the graphite obtained out of this anthracite.

Only industrial experience may decide which method is more efficient: graphitization of finely-ground anthracite with increased power consumption for polishing or graphitization of coarse material, which involves an increase of power consumption in furnaces and difficulties during grinding.

The observations just described are essential for the selection of raw materials for production, and lead to the following practical conclusions.

For the immediate future, it is suggested to use Donets anthracites possessing 5-6% ash content, minimum content of volatiles and having a thinly-laminated structure.

Design of a plant should include equipment for fine grinding of initial anthracites and a high-power polishing installation for the final product.

Rational design of a graphitizing furnace. The graphitizing furnace is the basic unit of equipment for producing artificial graphite, and its heating is the chief production operation. All other equipment and operations may be considered as secondary; their selection or design depend entirely on requirements set forth by the furnace and its features. However, up to the present, lack of a rational method for designing a graphitizing furnace creates considerable difficulties in the exploitation and planning of new installations.

The general operational principles of a graphitizing furnace are described in detail in the publication "Technology of Graphite" (1935), by V. S. Veselovskiy. The same publication describes an old furnace designing method based on empirical data and applicable only to furnaces closely related by their dimensions, heating rate and character of loading.

Furnaces of the Acheson type with accelerated heating are most convenient for manufacturing artificial graphite in powder form. Accelerated heating requires a certain increase in power of electrical equipment over the standard model, but this increase is entirely compensated by the increased efficiency factor of the furnace. In addition, selection of a fast furnace is predetermined by the requirements of Main Electric Power Administration (Glavenergo) to use electric power according to a compulsory schedule only 16-20 hours per 24 hour period.

The Acheson furnace is presented schematically in Figure 3. In the process of charging, first the bottom is covered with a fill of coke-sand mixture. After installing shields between the electrodes, powder to be processed is poured into the space formed between shields, being surrounded on the sides and top by the same coke-sand fill. To keep this fill in place, a temporary single-brick wall is to be built on both sides.

The core, made of thin electrodes or chunks of coal, is installed along the axis of the working space during furnace charging. This core serves for initial concentration of maximum current density along the axis, thus securing symmetrical heating of the furnace. As the powder to be graphitized is heated its electric conductivity increases proportionally and the cylinder of maximum current density (and maximum temperature) gradually expands. The process is completed when this cylinder occupies almost the entire volume of powder.

Hence, there are the following zones in the furnace at the moment of termination of heating: the grade I zone, occupying the middle portion of the working space; it is surrounded by the layer of grade III product, i.e. coal powder not heated to the temperature of complete graphitization; further, there is a sintered "carborundum" crust and, finally, the remaining portion of fill.

Let us assume that the material for graphitization is anthracite with 90% of 200-mesh (0.075 mm) component, having a loose weight of 0.75, ash content 6% and containing 5% of volatiles and moisture.

The amount of heat, transferred to the furnace during its heating, is distributed as follows: 1) heating grade I product - useful power; 2) heating grade III; 3) formation of carborundum sintered crust; 4) heating the filling material; 5) heating the furnace brick work; 6) heat loss through electrodes; 7) heat emission into space surrounding the furnace; 8) heat carried away by gases; 9) sublimation of ash admixtures.

These quantities of heat depend on the capacity of the furnace, its dimensions and distribution of temperatures. The latter depends on the rate of heating.

Heat emission into surrounding space and also heat carried out by gases and heat used for sublimation of ash admixtures may be ignored since their quantities are comparatively small and these losses are compensated through heat generated by combustion of gases and carbon.

The furnace volume is determined by a given productive capacity: M kg of grade I product per single campaign. This product occupies in the furnace working space a cylindrical volume:

$$V = \frac{M}{d} = \pi r^2 l$$

Around this volume, there are cylindrical layers of grade III product 1 dm thick, carborundum crust 0.5 dm thick and filling material of 2 dm thickness.

In addition, the brickwork, in the shape of a bath tub, has to be taken into consideration in heat calculations. Since the amount of heat absorbed by the brickwork is comparatively small, it may be assumed, for the sake of simplicity in calculations, that the brickwork has a shape of $^2/_3$ cylindrical layer 2.5 dm thick.

Volumes of layers:	Densities:
$V_{grade\ III} = V(\frac{2}{r} + \frac{1}{r^2})$	$d_{III} = 0.75$
$V_{SiC} = V(\frac{1}{r} + \frac{1.25}{r^2})$	$d_{SiC} = 1.0$
$V_{fill} = V(\frac{4}{r} + \frac{10}{r^2})$	$d_{fill} = 1.0$
$V_{brick} = V(\frac{3.33}{r} + \frac{15.8}{r^2})$	$d_{brick} = 1.2$

Calculation of the heat amount required for each layer, may be done if a certain definite distribution of temperatures in the furnace is given. The following distribution of temperatures may be assumed on the basis of experiments heretofore carried out: grade I product - 2400°; grade III - from 2200 to 1800°, average 2000°; carborundum crust - from 1800 to 1600°, average 1700°; filling material - in average 800°; brickwork - in average 200°.

Power consumption per 1 kg in various parts of the furnace under these conditions will be:

For grade I product:

1.	Heating to 2,400°:	$2{,}400 \cdot 2.26 \cdot 10^3$	$= 5{,}430 \cdot 10^3$ joules
2.	Evaporation of voltiles and moisture	$110 \cdot 2680$	$= 294.8 \cdot 10^3$ "
3.	Reduction of ash oxides		$500 \cdot 10^3$ "
	Total		$6{,}225 \cdot 10^3$ joules

or $\frac{6.225 \cdot 10^3}{3.600 \cdot 10^3} = 1.73$ kwh/kg

For grade III product:

1. Heating to 2,000°: $2{,}000 \cdot 2.0 \cdot 10^3 = 4{,}000 \cdot 10^3$ joules
2. Evaporation of volatiles $110 \cdot 2.680 = 300 \cdot 10^3$ "
3. Reduction of ash oxides $= 500 \cdot 10^3$ "

Total $4{,}800 \cdot 10^3$ joules =

= 1.33 kwh/kg

For the layer of carborundum crust:

1. Heating to 1,700°: $1{,}700 \cdot 0.8 \cdot 10^3 = 1{,}280 \cdot 10^3$ joules
2. Evaporation of volatiles (3%) $30 \cdot 2{,}680$ $80.5 \cdot 10^3$ joules
3. Reduction of SiO_2 into SiC. This very important item unfortunately can not be calculated with desirable accuracy, since it is unknown what portion of SiO_2 transforms into SiC in this layer.

It may be assumed that only half the crust transforms into carborundum, and power consumption per kg of crust is $9{,}119^{02} \cdot 10^3$ joules = 2.53 kwh/kg.

This value however must be considered as maximum, because filling material already previously used, and partly transformed into carborundum, ordinarily is utilized in production. Further, 2.0 kwh/kg is accepted for calculations.

For remaining filling material:

1. Heating to 800°: $800 \cdot 1.25 \cdot 10^3 = 1000 \cdot 10^3$ joules
2. Evaporation of volatiles (3%) $30 \cdot 2{,}680 = 80.5 \cdot 10^3$ joules

Total $1080 \cdot 10^3$ joules =

= 0.3 kwh/kg

For Brickwork:

1. Heating to 200°: $200.1 \cdot 10^3 = 200 \cdot 10^3$ joules = 0.0555 kwh/kg

Heat quantities, consumed for heating the furnace parts, will be expressed as:

$$Q_I = V \cdot 0.75 \cdot 1.73;$$

$$Q_{III} = V\left(\frac{2}{r} + \frac{1}{r^2}\right) \cdot 0.75 \cdot 1.33;$$

$$Q_{SiC} = V\left(\frac{1}{r} + \frac{1.25}{r^2}\right) 1.2;$$

$$Q_{fill} = V\left(\frac{4}{r} + \frac{10}{r^2}\right) 1 \cdot 0.3$$

$$Q_{brick} = V\left(\frac{3.33}{r} + \frac{15.8}{r^2}\right) 1.2 \cdot 0.0555$$

To obtain total heat consumption, heat loss through electrodes (Q_e) must be added to the above calculated values. Precise calculation of Q_e can not be presently performed. Obviously, its value must be proportional to the base of a cylinder representing the shape of the grade I product. Average temperature for the electrode portion, protruding from the brickwork, may be assumed equal to 1,500°. Heat quantity required for heating to this temperature is:

$$\frac{1{,}500 \cdot 1.75 \cdot 10^3 \cdot 4.0\, \pi r^2 \cdot 1.2}{3{,}600} = 5.8\, \pi r^2$$

However, heat is partially generated in electrodes due to current passing through them, and, at the present time, it is absolutely impossible to determine what portion of heat is received by electrodes from the working space of the furnace. But this item can not be ignored since it greatly affects the optimum ratio between length and radius of the furnace. It may be approximately assumed:

$$Q_e = 3\pi r^2$$

Adding all items of heat consumption, we obtain:

$$Q = V\left(1.3 + \frac{5.4}{r} + \frac{7.5}{r^2}\right) + 3\pi r^2$$

This equation expresses the heat quantity absorbed by the furnace as a function of furnace capacity and radius of the grade I product cylinder. It attains a minimum at very large radii, i.e. for furnaces considerably shorter than those used for graphitization, because electric losses increase with a decrease of furnace length.

This equation may be verified by comparison with the results of experiments conducted at "Elektrougli" plant.

Campaign No 14 (1932)

Power consumption in the generator circuit amounts to 3,474 kwh. Assuming losses in electrodes and bus-bars equal to 20%, we obtain 2,780 kwh as a value for power supplied to the furnace.

Calculation according to the equation gives Q = 2894.5 kwh.

Better agreement of the theory with experiment can not even be expected since, first, a number of simplifications was accepted in the development of the formula and, on the other hand, power consumption in the furnace was not determined by direct measuring but was calculated from operational conditions of the furnace.

Campaign No 2 (1936)

The furnace received 2,810 kwh

Calculation according to the formula gives Q = 2,357 kwh.

Campaign No 3 (1936)

The furnace obtained 1,910 kwh.

The formula gives Q = 1,862 kwh.

Agreement for these campaigns is also satisfactory. Reduced value for campaigns of 1936 is due to very high specific power consumption, which should be considerably lower at proposed rapid heating.

The formula just developed shows that the minimum power consumption is obtainable at large radii, i.e. for very short furnaces. With lengthening of the furnace, power consumption rapidly increases. But at the same time, the furnace resistance also increases, causing more favorable distribution of electric power in the low-voltage circuit. Thus, with increase of the furnace length, the thermal efficiency factor $\frac{Q_I}{Q}$ decreases, but the electric efficiency $\frac{Q}{A}$ increases. Consequently, such optimum dimensions of furnace must exist when the total efficiency

$$\eta = \frac{Q_I}{A} = \frac{Q_I}{Q_I + Q + A_e} = \frac{1}{1 + \frac{Q}{Q_I} + \frac{A_e}{Q_I}}$$

is at its maximum which gives a rational basis for determining the dimensions of the furnace by its given capacity.

$$\frac{Q}{Q_I} = \frac{4.15}{r} + \frac{5.77}{r^2} + 2.3\frac{\pi r^2}{V},$$

where

$$Q_n = Q_{III} + Q_{SiC} + Q_{fill} + Q_{brick} + Q_e, \quad \frac{A_e}{Q_I} = \frac{A_e}{Q} \cdot \frac{Q}{Q_I}.$$

$$\frac{A_e}{Q} = \frac{R}{Rf} = \frac{l_e \rho_e S_f}{l \rho_f S_e} = \frac{\rho_e}{\rho_f}\frac{S_f}{S_e} l_e \frac{\pi r^2}{V}$$

where R_e is resistance of electrodes; R_f - resistance of furnace; l_e - length of two electrode packs; ρ_e - effective specific resistances of electrodes;

S_f - elective cross-sections of furnace; S_e - effective cross-sections of electrodes. Further, it is necessary to find from experiment a numerical value for $\frac{\rho_e}{\rho_f}$ and set a required ratio $\frac{S_f}{S_e}$

For campaign No 2 of 1936

$$\frac{\rho_e}{\rho_f} = \frac{A_e S_e l}{Q S_f l_e} = 0.306$$

For campaign No 3 of 1936 $\frac{\rho_e}{\rho_f} = 0.278$;

$\frac{S_f}{S_e}$ for campaign No 2 = 0.9 and for No 3 = 0.75

Assuming $l_e = 40, \frac{\rho_e}{\rho_f} = 0.3, \frac{S_f}{S_e} = 0.8$ and substituting these values into the previous formulae, we obtain:

$$\frac{A_e}{Q} = 0.3 \cdot 0.8 \cdot 40 \frac{\pi r^2}{V} = 9.6 \frac{\pi r^2}{V}$$

$$\frac{Q}{Q} = 1 + \frac{4.15}{r} + \frac{5.77}{r^2} + 2.3 \frac{\pi r^2}{V}$$

Substituting these results into the formula for total efficiency, we obtain:

$$\eta = \frac{1}{\left(1 + 9.6\frac{\pi r^2}{V}\right)\left(1 + \frac{4.15}{r} + \frac{5.77}{r^2} + 2.3\frac{\pi r^2}{V}\right)}$$

On the basis of these works, the author, jointly with I. A. Shapiro and D. A. Smirnov compiled design requirements for planning a plant for artificial graphite.

Site for building this plant was selected in the vicinity of Shakhty in the eastern part of Donbas. The location secures supply of raw materials and electric power and is near consumers. This plant will be the first experiment of producing

Acheson graphite in the USSR and, therefore, should have the character of an experimental plant, having a productive capacity not very high. For the beginning, 3,000 tons annually was suggested, i.e. 12 tons per working day.

Donets anthracite of finely-laminated structure with an ash content of 5-6% and with moisture and volatiles not more than 6% has to serve as raw material for production.

During furnace charging, a cylinder of coarse anthracite and grade III product from previous campaigns should be installed along the axis of working space. This cylinder must be packed around with fine powder of anthracite, followed by filling material.

Average power consumption may be assumed equal to 5 kwh per kg of product; from this daily power consumption amounts to 60,000 kwh. Total power at 16 hours of daily operation of all furnaces equals 3,750 kw.

To secure uniform load on three-phase line, it is necessary to have at least 3 furnaces. Subsequently, average power of each furnace is 1250 kw and capacity - 4,000 kg of product or 5340 kg of initial material with loose weight of 0.75; useful consumption of power for a single campaign $Q_I = 4{,}000 \cdot 1.73 = 6{,}920$ kwh.

Substituting these data into the formula for rational calculation of the furnace we obtain its efficiency factor at various lengths. Results of calculations are represented graphically in Figure 4. The graph shows maximym efficiency for radii of the working space (r) from 6 to 7.5 dm. This corresponds to a distance between furnace electrodes from 3 to 5 meters. The upper limit should be used, since with increase of the length of furnace its effective resistance increases faster than reactance and, consequently, cos φ increases also.

For example, the following approximate values are obtainable at the end of furnace heating when 3 and 5.5 m are working space lengths:

	Length	
	3m	5.5m
Effective resistance of working space (mΩ)	0.72	2.43
Effective resistance of electrodes	0.23	0.42
	0.95	2.85
Reactance of furnace	1.44	2.24
Cos	0.55	0.79

In addition, in the case of the longer furnace the current in the low-voltage circuit is lower and, consequently, electric losses are smaller.

Since the bottom layers of anthracite in the furnace charge are under higher pressure than the top layers, they have lower resistance, creating irregularities in the furnace heating process. To eliminate this effect, the working space cross-section must be given an elliptical shape.

The author's measurements demonstrated that the effective specific resistance of electrode packs in existing graphitizing installations is about 0.015 Ω/cm, i.e. its value reaches $^1/_3$ the resistance of the furnace working space, while the real resistivity of electrodes does not exceed 0.006-0.008 Ω/cm. The usual ratio between cross-sections of the working space and electrodes, about 0.8, causes high losses of power in electrodes (about 25%). These losses must be decreased by increasing the cross-section of electrode packs and by the rational construction of electrode holders, which secures uniform current distribution along the pack cross-section. In the USA for this purpose each electrode in the pack is equipped with a nipple through which current is fed into electrode.

Two layouts were developed by the author for realization of the production process: the first layout is the usual arrangement with stationary furnaces installed in a single large furnace shop, the second - with furnaces on carriages movable to the reducing transformers for heating.

Second variant has more advantages due to lower investment into a building, and from the operational viewpoint, since it permits maximum mechanization of this labor-consuming and difficult manufacturing process.

Furnace unloading is the most difficult operation in the production of artificial graphite. It may be mechanized with the aid of a pneumatic system in cases of fine-grained initial material since the latter does not become cemented during graphitization and may be easily sucked out of the furnace. Pneumatic unloading in addition to mechanization of the process, has another advantage: possibility for unloading a hotter furnace.

Artificial graphite must be released from the plant in the finely ground ($<$ 250-mesh) state and thoroughly polished. Ball mills are most suitable for finishing. Mills of periodic action were tested up to the present. Time of treatment increases with increase in the fineness of product. At 1:2 ratio of the product weight to the weight of balls the polishing process takes about 5 hours.

It is possible also to use mills of continuous action connected with an air classifier.

Schematically the equipment in this case may be presented as follows. The pneumatic system feeds product from the furnace into a cyclone separator under which the bunkers of ball mills are installed. Air and fine dust go to a dust chamber and graphite from bunkers enters ball mills; on the exit from mills it is sucked into another cyclone which yields final product, returning insufficiently fine graphite to the mill. The air from this cyclone also goes to a dust collector.

A special ball mill with classifier should be provided for grinding graphites of higher grade obtained from the core of the graphitizing furnace.

Blast Furnace Graphite

On cooling of molten pig iron, tapped from a blast furnace, a certain part of the dissolved carbon crystallizes out as graphite which, due to its low specific gravity, floats on the surface of the molten metal.

During the operation of cleaning the laddles after metal pouring, this graphite is mixed with sand from the lining and coke used on top of the molten metal for its protection against oxidation. Usually these scrapings are returned to the blast furnace.

The total quantity of graphite formed in this way at large metallurgical plants is fairly considerable. The largest amount of graphite is yielded by foundry pig iron. Open hearth pig irons with low content of silicon give just a little of graphite.

Foundry pig iron may give an amount of graphite equal to 0.4 -0.5% of its own weight. Since smelting of this product in the Union reached 3.5 million tons in 1936, the total quantity of blast furnace graphite is estimated at 15,000 tons.

Of course, complete utilization of this graphite is impossible; its collection may be profitably organized only at large plants; there are some losses of graphite during the transportation and pouring of pig iron. Preliminary considerations show that not more than 50% of total blast-furnace graphite may be utilized in the near future. But even this amount is so large that, for example, it exceeds the entire quantity of high grade graphite produced by all graphite-dressing plants of the Soviet Union. In any case, it deserves to be paid attention.

The first significant experiments for graphite extraction out of blast-furnace scrap were carried out in 1934 by All-Union Graphite-Corundum Trust, Azov Steel and IMS. These experiments showed that scrap from pouring laddles, passing through a 5 mm sieve, contains about 20% graphite. The insignificant graphite concentration, remaining in the coarser classes of scrap, may be ignored.

Fine grade ($<$ 5mm), besides graphite, contains pieces of metallic iron (shot), iron oxides and sand. Early attempts for further graphite concentration by the flotation method failed because pig iron shot clogged classifier and flotation equipment.

V. S. Veselovskiy and M. A. Eygeles, member of the IMS, suggested air separation for coarse-grained admixtures. Experiments, conducted in 1934, showed the possibility of obtaining a product with 20-25% ash content. Flotation of this product has no difficulties, giving a concentrate with 5-8% ash content.

On the basis of these observations, the All-Union Graphite-Corundum Trust equipped the experimental installation at the Mariupol' Graphite Plant. Simultaneously the IMS initiated development of a rational method for extraction of blast-furnace graphite, and plant design. These works were executed by M. A. Eygeles. However further practical development was interrupted by lack of money, in spite of the fact that tests of blast-furnace graphite at the Lugansk Crucible Plant gave very favorable results. To a considerable extent, this failure may also be attributed to the fact that organization of collection of graphite scrap requires considerable flexibility and mobility, i.e. qualities which are not at all in the character of such a cumbersome establishment as the All-Union Graphite-Corundum Trust.

Activity on the subject was resumed by the IMS in 1936 in connection with a general sharp increase in the scarcity of graphite. I. A. Shapiro, member of the IMS investigated possibilities for the collection of graphite at the largest blast-furnace plants. This investigation completely corroborated the favorable outlooks for the utilization of blast-furnace graphite. He selected and delivered to Moscow 4 large samples of screened graphite scrap weighing several tons each. The main purpose of these investigations was not a study of methods for scrap processing, but preparing large samples of concentrate for testing in various industries, first of all in cell, pencil and graphite-ceramic production.

Products with 16-26% ash content were obtained by air separation. Since the flotation method was not applicable, these products were ground and treated with hydrochloric acid. The final product with 5-6% ash content was tested in pencil and cell production. Preliminary experiments of manufacturing galvanic cells showed good results. Experiments on a bigger scale were intended in 1937. Mechanical treatment of the final product, having the same significance for blast-furnace graphite as for Acheson graphite, should be studied in particular.

On the whole, the situation with blast-furnace graphite at the present time appears to be as follows. First of all, there is no doubt that it deserves consideration from the industrial viewpoint. Organization of its collection gives considerable profit and is fully practicable.

1. It will permit utilization of the waste products of blast-furnace industry.

2. The industry will obtain valuable and scarce material at the rate of 4,000 tons per year, valued at approximately 4 million roubles.

3. In the case of unforeseen need for graphite it will give a new supply source, organization of which is comparatively simple and may be readily developed.

4. Considerable economy in investments may be achieved since organization of the collection and concentration of blast-furnace graphite entails considerably smaller expenses than does the construction of plants for dressing natural graphitic ores.

5. Territorially, the manufacture of blast-furnace graphite coincides nearly with sites of its consumption.

There are no doubts in respect to the feasibility of blast-furnace graphite beneficiation, but there still remains the necessity of clarifying some details and, what is particularly essential, fields of application.

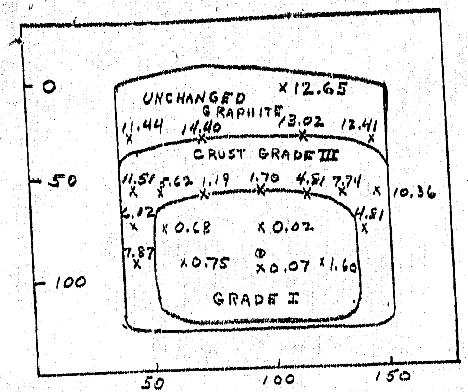

Fig 1: Topogram of Ash Content Distribution (Campaign No 1).

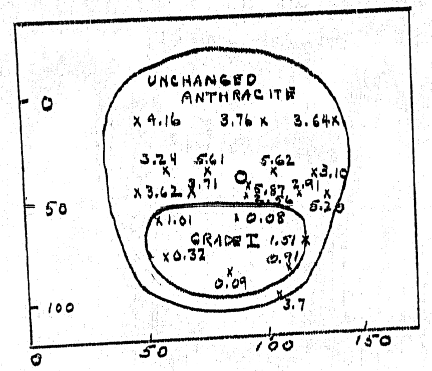

Fig 2: Topogram of Ash Content Distribution (Campaign No 2).

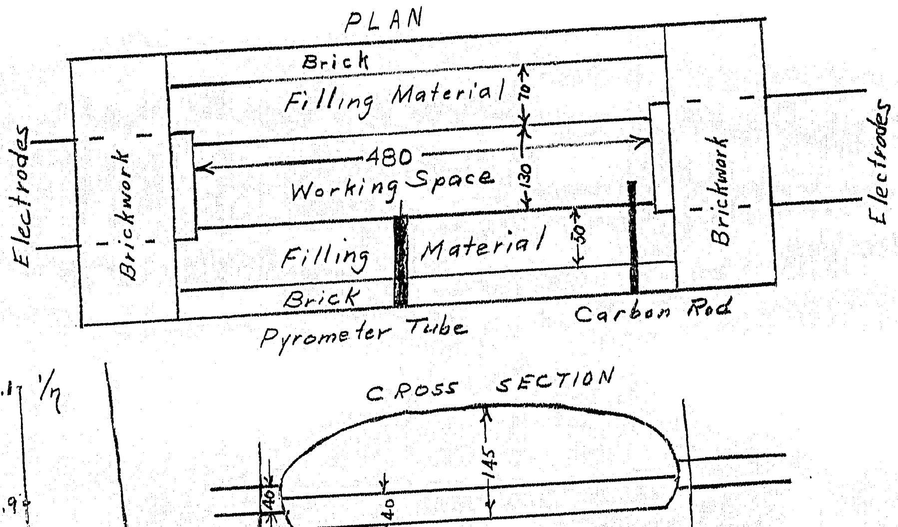

Fig 3: Layout of Graphitizing Furnace.

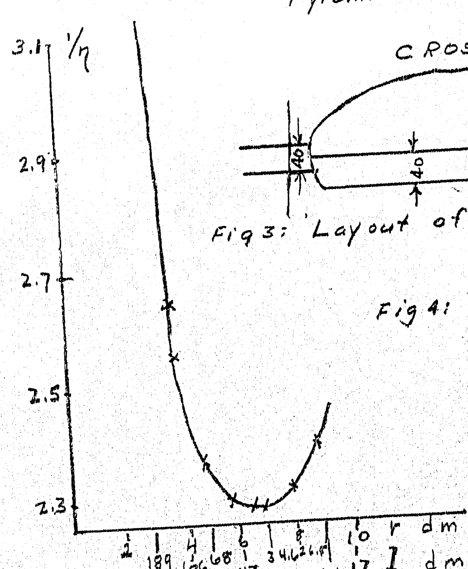

Fig 4: Relationship Between the Efficiency of the Graphitizing Furnace and its Length at Constant Productive Capacity.